

## Silicon Isotope Separation by Infrared Free Electron Laser

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Silicon isotope separation through irradiation of  $\text{Si}_2\text{F}_6$  using an infrared free-electron-laser (FEL) in the  $800\text{ cm}^{-1}$  and  $1000\text{ cm}^{-1}$  regions was examined. By controlling the laser wavenumber, preferential decomposition of  $\text{Si}_2\text{F}_6$  into  $\text{SiF}_4$  and  $\text{SiF}_2$ , enriched with  $^{29}\text{Si}$  and  $^{30}\text{Si}$  isotopes, was induced.

Key words: silicon, isotope separation, infrared free electron laser,  $\text{Si}_2\text{F}_6$ .

### 1. INTRODUCTION

Silicon consists of three stable isotopes:  $^{28}\text{Si}$ ,  $^{29}\text{Si}$  and  $^{30}\text{Si}$ . By controlling the isotope ratio, the thermal and nuclear properties of silicon can be improved [1]. A single crystal of  $^{28}\text{Si}$  is predicted to have a high thermal conductivity due to the absence of isotope scattering by other isotopes [2].  $\text{SiC}$  consisting of  $^{29}\text{Si}$  and  $^{30}\text{Si}$  is an attractive candidate as a material with low induced-radio-activation in high energy neutron irradiation environments [1]. Enrichment with  $^{30}\text{Si}$  is advantageous in the production of n-type semiconductor doped with phosphor by neutron transmutation doping [3]. To realize such isotopically controlled silicon, a practical isotope separation process is required. Laser isotope separation with infrared multi-photon decomposition is very attractive method of producing a large amount of isotopes. Recently, silicon isotope separation of  $\text{Si}_2\text{F}_6$  with a  $\text{CO}_2$  infrared laser at  $1000\text{ cm}^{-1}$  was reported [4,5]. However,  $\text{Si}_2\text{F}_6$  has additional absorption peaks at around  $400$  and  $800\text{ cm}^{-1}$ . But, because of the limitations of  $\text{CO}_2$  laser emission lines, it is not obvious that it is possible to achieve isotope separation with  $\text{CO}_2$  laser at these other infrared wavenumbers, while the free electron laser is tunable to these wavenumbers.

In the present paper, we study silicon isotope separation from  $\text{Si}_2\text{F}_6$  by using an infrared free electron laser (FEL), in particular in the  $800$  and  $960\text{ cm}^{-1}$  regions.

### 2. EXPERIMENTAL

The  $\text{Si}_2\text{F}_6$  used in these experiments was produced by fluorination of  $\text{Si}_2\text{Cl}_6$  with  $\text{ZnF}_2$ .  $\text{Si}_2\text{F}_6$  contained a trace of  $\text{SiF}_4$  impurities. After purification of the  $\text{Si}_2\text{F}_6$ , the final  $\text{SiF}_4$  impurity was  $0.06$  and  $0.09\text{ vol}\%$  for two lots of  $\text{Si}_2\text{F}_6$ .

Figure 1 shows a schematic of the experimental apparatus. The infrared FEL at the FOM Institute for Plasma Physics in the Netherlands was used as a light source. The wavenumbers of the FEL were set at  $786$ - $814$  and  $950$ - $972\text{ cm}^{-1}$ . A laser pulse consisted of  $10\mu\text{s}$  macropulse with a  $8.33$ - $10\text{Hz}$  repetition frequency composed of  $1\text{ ns}$  micropulses with a repetition rate of  $1\text{ GHz}$ . The beam diameter was  $2\text{ mm}$  and the laser energy was  $12$ - $27\text{ mJ}$  in front of the reaction cell.

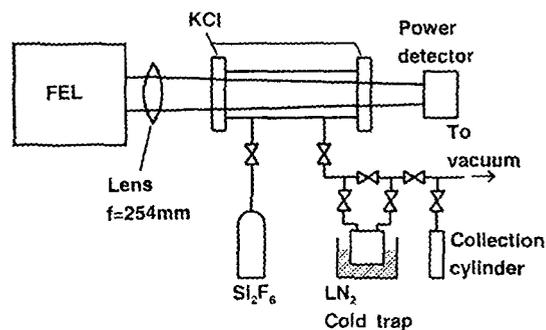


Fig. 1 Schematic of experimental apparatus.

The laser beam was focused by a ZnSe lens and introduced into a reaction cell. The reaction cell was a cylindrical Pyrex glass tube,  $100\text{ mm}$  long,  $10\text{ mm}$  in inner diameter, and equipped with KCl windows at both ends. The flow rate and pressure of  $\text{Si}_2\text{F}_6$  were set at  $8.3\text{ mm}^3\text{s}^{-1}$  and  $13.3\text{ Pa}$ . The irradiation was performed for  $300\text{ s}$  at room temperature and decomposed the  $\text{Si}_2\text{F}_6$  into  $\text{SiF}_4$  gas and  $\text{SiF}_2$  polymer. Both product  $\text{SiF}_4$  and residual  $\text{Si}_2\text{F}_6$  were captured with a liquid nitrogen cold trap and then separated into each component through low temperature distillation. The isotope ratios were determined by using a quadrupole mass spectrometer from the relative ion intensities of the isotope species.

### 3. RESULTS AND DISCUSSION

Figure 2 shows the infrared absorption spectrum of natural  $\text{Si}_2\text{F}_6$  in the region of  $700$ - $1100\text{ cm}^{-1}$ . There are strong absorption peaks at  $990$  and  $820\text{ cm}^{-1}$  [6,7]. The peak at  $990\text{ cm}^{-1}$  is due to an asymmetric stretching vibration, and the peak at  $820\text{ cm}^{-1}$ , to a symmetric vibration of the Si-F bond. Since the natural abundance ratio of silicon is  $^{28}\text{Si}:^{29}\text{Si}:^{30}\text{Si} = 92.23:4.67:3.10$ , these peaks are attributed to the vibration of  $\text{Si}_2\text{F}_6$  molecules containing  $^{28}\text{Si}$ . Though the spectral lines for  $^{29}\text{Si}$  and  $^{30}\text{Si}$  are not apparent in this figure, they should be present around  $990$  and  $820\text{ cm}^{-1}$  [8]. Since the emission of the FEL covers these ranges, infrared multi-photon decomposition of  $\text{Si}_2\text{F}_6$  molecules into products

containing  $^{29}\text{Si}$  and  $^{30}\text{Si}$  can occur effectively by selecting the appropriate wavenumber. The reaction producing  $\text{SiF}_4$  by infrared multi-photon decomposition is assumed to be

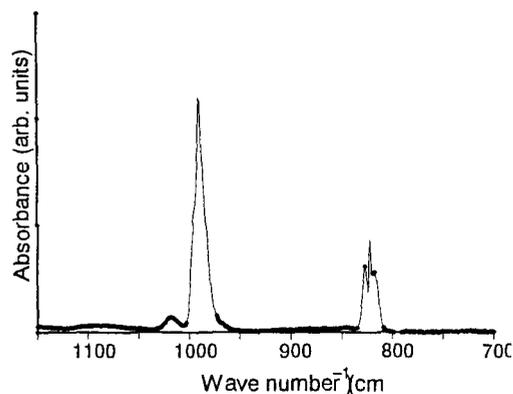
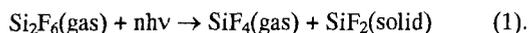


Fig. 2 Infrared spectrum of  $\text{Si}_2\text{F}_6$  around 700 to 1100  $\text{cm}^{-1}$ .

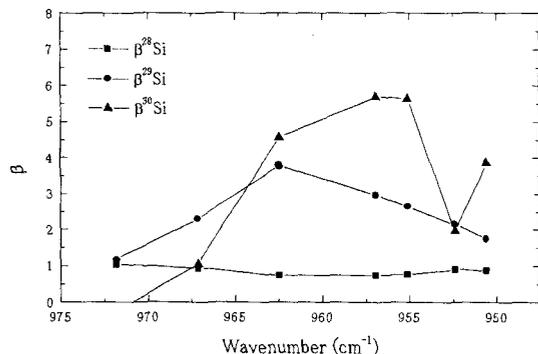


Fig. 3 Enrichment factor of Si in the product  $\text{SiF}_4$  as a function of wavenumber around 960  $\text{cm}^{-1}$ .

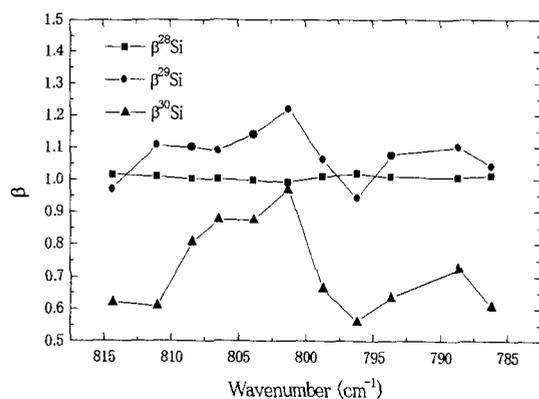


Fig. 4 Enrichment factor of Si in the product  $\text{SiF}_4$  as a function of wavenumber around 800  $\text{cm}^{-1}$ .

Figure 3 shows the enrichment factor of Si in the product  $\text{SiF}_4$  as a function of wavenumber around 960  $\text{cm}^{-1}$ , where the enrichment factor  $\beta^i$  is defined as

$$\beta^i = (\text{product } ^i\text{SiF}_4)/(\text{natural } ^i\text{SiF}_4) \quad (2).$$

When  $\text{Si}_2\text{F}_6$  was irradiated around 950-972  $\text{cm}^{-1}$ , the product  $\text{SiF}_4$  was enriched with  $^{29}\text{Si}$  and  $^{30}\text{Si}$ .  $\beta^{29}$  has a peak value of 3.8 at 962.5  $\text{cm}^{-1}$  and  $\beta^{30}$ , of 5.7 at 956.9  $\text{cm}^{-1}$ . These  $\beta$  values show that the product  $\text{SiF}_4$  is composed 17.7% of  $^{29}\text{Si}$  and 17.7% of  $^{30}\text{Si}$ .

Figure 4 shows the enrichment factor of Si isotopes in the product  $\text{SiF}_4$  as a function of wavenumber around 800  $\text{cm}^{-1}$ .  $^{29}\text{Si}$  was enriched in the product  $\text{SiF}_4$  between 786-814  $\text{cm}^{-1}$ .  $\beta^{29}$  has a peak value of 1.2 at 801.3  $\text{cm}^{-1}$ , while  $\beta^{30}$  is smaller than 1 for all wavenumbers. The difference of  $\beta^{30}$  between the 960  $\text{cm}^{-1}$  region and the 800  $\text{cm}^{-1}$  region is probably due to the difference of the vibration modes of the  $\text{Si}_2\text{F}_6$  molecules in the two regions.

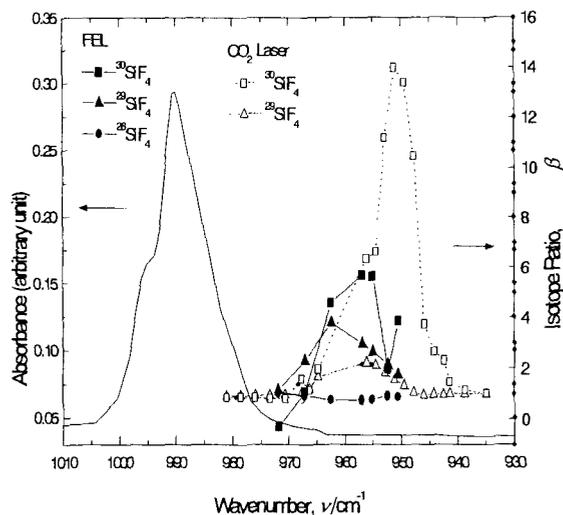


Fig. 5 The infrared spectrum of  $\text{Si}_2\text{F}_6$  (right axis), the FEL enrichment factors, and the  $\text{CO}_2$  laser enrichment factors.

Figure 5 shows the infrared spectrum of  $\text{Si}_2\text{F}_6$  (right axis), the FEL enrichment factors, and the  $\text{CO}_2$  laser enrichment factors. The FEL data are the same as those in Figure 3, and the  $\text{CO}_2$  laser data are from our previous study that was performed under the same experimental conditions but using a  $\text{CO}_2$  pulse laser [9]. Since there is only a small emission gain for the  $\text{CO}_2$  laser at 956-965  $\text{cm}^{-1}$ , the pulse energy was not large enough to decompose the  $\text{Si}_2\text{F}_6$ , so there is no data in that region, while the FEL can emit continuously in that region, producing a  $\beta^{29}$  enrichment peak at 962  $\text{cm}^{-1}$ . Compared to  $\text{CO}_2$  laser results, the enrichment factor of FEL is relatively low. This can be attributed to two features of the FEL pulse: the spectral width,  $\delta\lambda/\lambda$ , and the macropulse length. The spectral width and the macropulse length of the FEL are 1-2% and 10  $\mu\text{s}$

respectively, whereas those of the CO<sub>2</sub> laser are 0.003% and 104 ns, respectively. The broad spectral width and long pulse width probably contribute to the lower isotope selectivity of the FEL.

#### 4. CONCLUSION

The isotope separations of <sup>29</sup>Si and <sup>30</sup>Si were made using the isotope selective infrared multi-photon decomposition of Si<sub>2</sub>F<sub>6</sub> by infrared the FEL irradiation.

A relatively high enrichment of <sup>29</sup>Si and <sup>30</sup>Si in the product SiF<sub>4</sub> was observed for laser irradiation in the range 950-972 cm<sup>-1</sup>.

For the first time, enrichment of <sup>29</sup>Si at around 800 and 960 cm<sup>-1</sup> has been achieved, by using an FEL rather than a CO<sub>2</sub> laser.

The low enrichment factor for the FEL compared to the CO<sub>2</sub> laser is probably due to the broader spectrum width and longer pulse width of the FEL.

#### REFERENCES

- [1] T. Noda, *Kinzoku* **7**, 32(1993).
- [2] P. G. Klements: *Int. J. Thermophysics*, **2**, 323(1981).
- [3] M. Tanenbaum, et al. *J. Electrochem. Soc.*, **108**,171(1961).
- [4] M. Kamioka, et al. *J. Phys. Chem.*, **90**, 5727(1986).
- [5] S. Arai, et al. *Appl. Phys.*, **B53**, 199(1991).
- [6] V. Tosa, et al. *Vib. Spectrosc.*, **29**, 631(1994).
- [7] V. Tosa, et al. *J. Mol. Struct.*, **410**, 411(1997).
- [8] L. Halonen, *J. Mol. Spectrosc.*, **120**, 175(1986).
- [9] H. Suzuki, et al. *J. Jpn. Int. Met.* **61**,145(1997).

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